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研究论文

不同锆源对 $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ 整体式三效催化剂性能的影响

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- 摘要
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摘要 分别以 $ZrOCO_3$, $ZrO(NO_3)_2$ 和 $Zr(NO_3)_4$ 为锆源采用双氧水氧化-共沉淀法合成了 $Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ 储氧材料, 并用湿法浸渍法制备了 $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ 三效催化剂, 用 X 射线衍射、拉曼光谱、N_2 吸附、X 射线光电子能谱、氢气程序升温还原和氧脉冲等技术对该催化剂进行了表征. 结果表明, 以 $ZrOCO_3$ 和 $ZrO(NO_3)_2$ 为锆源时所制催化剂上只能检测到单一的 CeO_2-ZrO_2 立方相, 而以 $Zr(NO_3)_4$ 为锆源制备的催化剂经老化后却能检测到四方相; 另外, 在模拟汽车尾气组分和工作条件下考察了整体型 $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.9}$ 催化剂的三效活性. 结果表明, 以 $ZrOCO_3$ 为前驱体制备的整体式 $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.9}$ 催化剂对 C_3H_8, CO 和 NO 具有最佳的催化活性. 认为 $ZrOCO_3$ 更有利于 Zr^{4+} 和 La^{3+}进入 CeO_2 晶格形成均一固溶体, 从而提高催化剂的高温稳定性和氧化还原性能.

关键词: 储氧材料 氧化共沉淀 热稳定性 氧化还原性能 三效活性

Abstract: Three types of $Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ mixed oxides were prepared by H_2O_2-assisted precipitation using $ZrOCO_3$, $ZrO(NO_3)_2$, and $Zr(NO_3)_4$ as precursors. The $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ three-way catalysts were prepared by the wet impregnation and characterized by the X-ray diffraction (XRD), Raman spectroscopy, nitrogen adsorption, X-ray photoelectron spectroscopy (XPS), temperature-programmed reduction (TPR), and oxygen storage capacity. XRD and Raman spectroscopy results showed that the use of $ZrOCO_3$ or $ZrO(NO_3)_2$ gave a CeO_2-ZrO_2 mixed oxide with a cubic phase of high thermal stability, while an extra t″-phase was observed in the aged $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ catalyst prepared using $Zr(NO_3)_4$. The activities of the three catalysts were evaluated using simulated exhaust gas under working conditions. The $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ catalyst prepared using $ZrOCO_3$ as precursor gave the best activities for C_3H_8, CO, and NO conversion. The use of $ZrOCO_3$ as precursor was better suited for the formation of Ce^{3+} and more favorable for the homogeneous insertion of Zr^{4+} and La^{3+} into the CeO_2 lattice, which resulted in better thermal stability and enhanced redox properties of the $Pd/Ce_{0.45}Zr_{0.45}La_{0.1}O_{1.95}$ catalyst.

Keywords: oxygen storage material, oxidation-coprecipitation, thermal stability, redox property, three-way activity

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